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Measurements have been made of the electrical and optical properties of electrical discharges in Mg-Xe mixtures at moderate current densities (50-100 A/cm^2) and high Xe pressures (1 to 5 atmospheres). Preliminary analyses indicate a rather low electron and excitation temperature resulting from unexplained large energy losses. Efforts to obtain the high excitation temperatures required for efficient, high power excimer laser operation will continue.

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20. Abstract (continued):

An experiment is being designed to provide effective rate coefficients for electron excitation and deexcitation of excited mercury atoms so as to provide data necessary for the modeling of the contribution of these states to the production of upper laser levels and to the growth of ionization and instabilities in discharge excited lasers. This experiment will use laser induced perturbation of the excited state densities and optical observation of the relaxation of excited state densities.

Measurements have been made of the absolute rate coefficients for electron excitation of the $C^3\Pi$ state of N_2, i.e., for the upper state of the uv N_2 laser. Absolute emission intensity measurements were made for mean electron energies between 1 and 7 eV and N_2 pressures from 2 to 20 torr. This data has been used to derive an improved set of electron excitation cross sections for N_2.

SEMIANNUAL REPORT

This Semiannual Report contains descriptions of work carried out under ONR Contract No. N00014-76-0123 and ARPA Order No. 2683-Amd. 8. It covers the period from 1 August 1978 to 31 January 1979. Section I is the Seminannual Report Summary while Sections II-III are more detailed descriptions of work carried out under the projects supported by this contract.

I. SEMIANNUAL REPORT SUMMARY

The projects being carried out under this contract are summarized below. More detailed discussions are given in Sections II-IV of this report.

(1) Metal Vapor-Rare Gas Discharges

The objective of this project is the evaluation of the potential of electrical discharges in high pressure metal vapor-rare gas mixtures for excitation of high power, high efficiency lasers operating at near visible wavelengths. Our work under this and the previous ARPA/ONR contract has provided stimulated emission coefficients for several metal vapor-rare gas excimers at visible or near visible wavelengths and has shown that electrical discharges with sufficiently high electron temperatures (>0.5 eV) should be an efficient way to produce these excimers. Our recent experiments under this contract have shown that the effective electron temperature in high power discharges in Na-Xe mixtures with Na densities between 10^{15} and 10^{16} cm^{-3} is too low to yield useful gain on the Na-Xe excimer transition. However, a model of these Na-Xe discharges has been used to extrapolate the discharge behavior to much higher Na densities, e.g., 3×10^{17} cm^{-3}, where the electron temperature and optical gain should be sufficient to yield a reasonably efficient laser device.

In an effort to find metal vapor-rare gas mixtures with better laser gain and electrical properties we began a study of the optical and electrical properties of electrical discharges in mixtures of Mg and Xe at high pressures. We find these discharges to have electrical properties similar to those of discharges in Na-Xe mixtures, e.g., a reasonably diffuse positive column with indications of a positive volt-ampere characteristic. From measurements of absolute intensities of spectral lines we find that the excitation temperature and, therefore, the apparent electron temperature is somewhat higher than for NaXe discharges at the same discharge current density.

(2) Electron Excitation Rates in Metal Vapors

The objective of this new project will be to determine effective rate coefficients for the electron excitation and deexcitation of excited mercury atoms for use in the modeling of the role of these excited atoms in electrically excited, high power lasers, e.g., mercury-halogen lasers. Because the halogens are chemical reactive these experiments are being carried out in mercury-rare gas mixtures. The electrical and optical properties of discharges in this mixture are being investigated under another contract. Once these discharges are characterized the present investigation will make use of laser perturbation techniques and fast transient analysis of the emitted radiation to obtain the desired rate coefficients. This means that the desired measurements will not be made until near the end of this contract.

(3) Electron Excitation of Metastable Atoms and Molecules

During this report period we completed determinations of the excitation rate coefficients for the $C^3\Pi_u$ state of N_2 in pure N_2. At the same time we began development of a new technique for the determination of excitation rate coefficients for the metastable states of the rare gases. The development of a new technique for the rare gas measurements was made necessary by the difficulty of the extraction of rare gas excitation rates from the measurements of $N_2(A^3\Sigma)$ metastable excitation rate coefficients in N_2-rare gas mixtures, i.e., in the range of mean electron energies available below electrical breakdown in our experiment most of the $N_2(A^3\Sigma)$ excitation was by direct electron excitation of the N_2 rather than by excitation transfer from Ar metastables. The determinations of rate coefficients for the electron excitation of the $N_2(C^3\Pi)$ state provided much inproved data for use in the optimization of the excitation of the upper state of the uv nitrogen laser as well as data essential to the proper analysis of the behavior of the higher energy electrons in N_2.

II. METAL VAPOR-RARE GAS DISCHARGES

Drs. W. L. Morgan (to 9/78), L. Schumann, A. Gallagher and A. V. Phelps

Experiments on a magnesium doped discharge in xenon have been performed during this report period on the apparatus that was used in the previous sodium-xenon[1] and thallium-xenon[2] experiments. The information gathered consists of identification of features in the spectrum of the discharge, values of electric field to gas density E/N, electron density n_e in the discharge, and magnesium excited state population densities.

Figure 1 shows measured spectral intensities for a typical magnesium-xenon discharge in the wavelength region from 2750 Å to 8500 Å. Most of the main features are identified in Fig. 1. Much of the power radiated is in the MgH bands at 5100 Å and 5550 Å. Because of the large vibrational spacing of the vibrational structure observed in the MgH energy levels, the vibrational structure of the band is readily observed. One notable exception is the vibrational structure at 3500 Å which we attribute to radiation from the $MgXe^+$ molecule, $(A^2\pi_{3/2} \rightarrow X^2\Sigma_{1/2})$. The radiation from the MgXe molecule in the first excited state is indicated by the dashed line in the region around 3500 Å. This radiation is associated with the $Mg(3^1P_1 \rightarrow 3^1S_0)Xe$ transition. There are also low level continuum in the regions near 4350 Å and 6000 Å which may be due to $Mg(3^3D_{1,2,3} \rightarrow 3^3P^0_{0,1,2})Xe$ and $Mg(4^3S_1 \rightarrow 3^3P_{0,1,2})Xe$, respectively.

As a general rule, the E/N's in magnesium-xenon discharges are about twice as large as in thallium-xenon[2] for a given current density. Thus the electron densities are about half of the values as in thallium-xenon since μN is only slightly different for the apparent electron temperature of 0.44 eV found for Mg-Xe at 100 A/cm^2 and the 0.5 eV found for Tℓ-Xe at the same current density J. Note that the higher E/N values cause the experimental power loss per atom, EJ/N,

to be higher in Mg-Xe than in Tℓ-Xe whereas the lower electron density in Mg-Xe means that the electron-xenon elastic recoil losses will be less than in Tℓ-Xe. This data shows the major source of power dissipation in the Mg-Xe discharge is not elastic recoil collisions of electrons with Xe. As in the case of Na-Xe our models fail to explain the major energy losses in these discharges.

The ratios of excited state density to statistical weight vs. excitation energy, i.e., the Boltzmann plot, for a typical Mg-Xe discharge is shown in Fig. 2. The spectrum used for this calculation was obtained under conditions which were about the same as those in Fig. 1. Figure 2 shows that excited state populations for the lower excited states are very near to those expected for thermal equilibrium at a temperature of 0.44 eV. The point at the ionization limit, [x], is an upper limit and is derived from the Saha equation at the ionization limit[1] by assuming $[Mg^+] = n_e = 8.5 \times 10^{14}/cm^3$. In magnesium there is an ion emission line at 2802 Å and measurements of the absolute intensity of this line yields $[Mg^+] = 4.8 \times 10^{13}/cm^3$ which corresponds to the lower point marked at the ionization limit. Having measured the magnesium ion density, one can determine the $MgXe^+$ density by charge balance. Thus $[MgXe^+] + [Mg^+] = n_e$ yields the ratio of $[MgXe^+]/[Mg^+] \simeq 15$.

An attempt will be made to extend these measurements and preliminary analysis of discharges in Mg-Xe mixtures so as to obtain higher electron temperature and more information regarding energy loss processes.

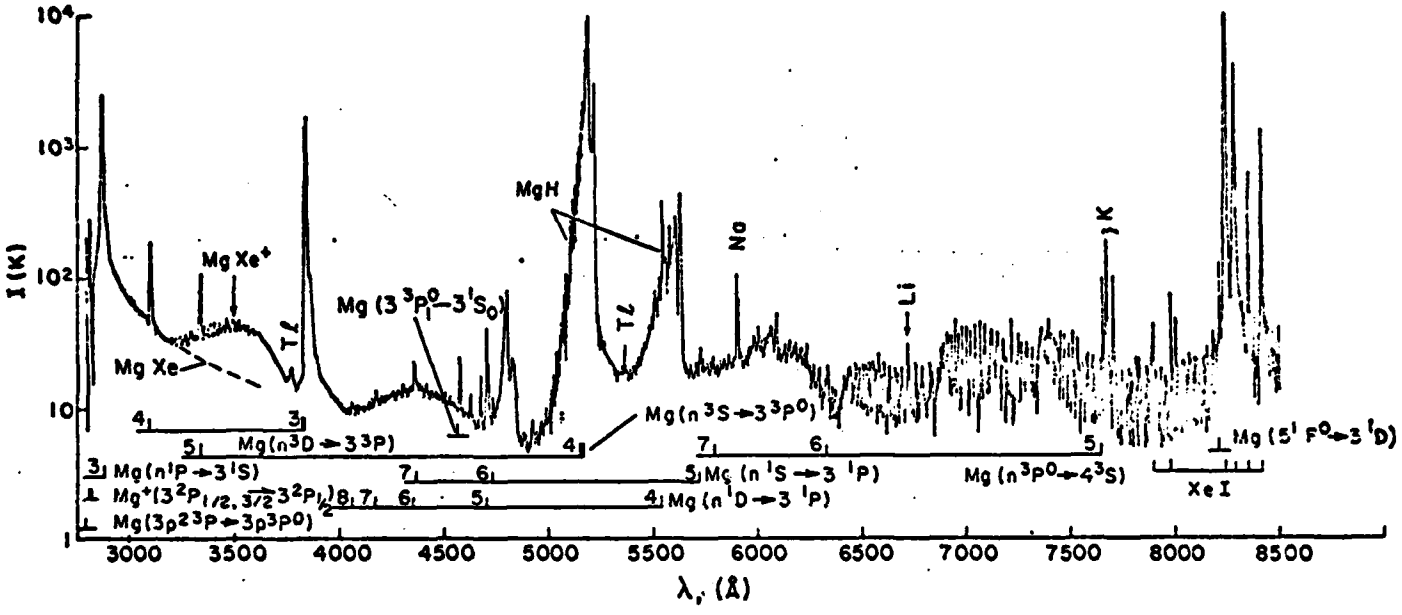

Fig. 1. Typical magnesium-xenon discharge spectrum $[Mg]_0 = 7.3 \times 10^{15}$ cm^3, $[Xe] = 2.53 \times 10^{19}$ cm^3, $n_e = 8.4 \times 10^{14}$/cm^3, E/N = 6.3×10^{-18} V-cm^2, J = 101.2 A/cm^2.

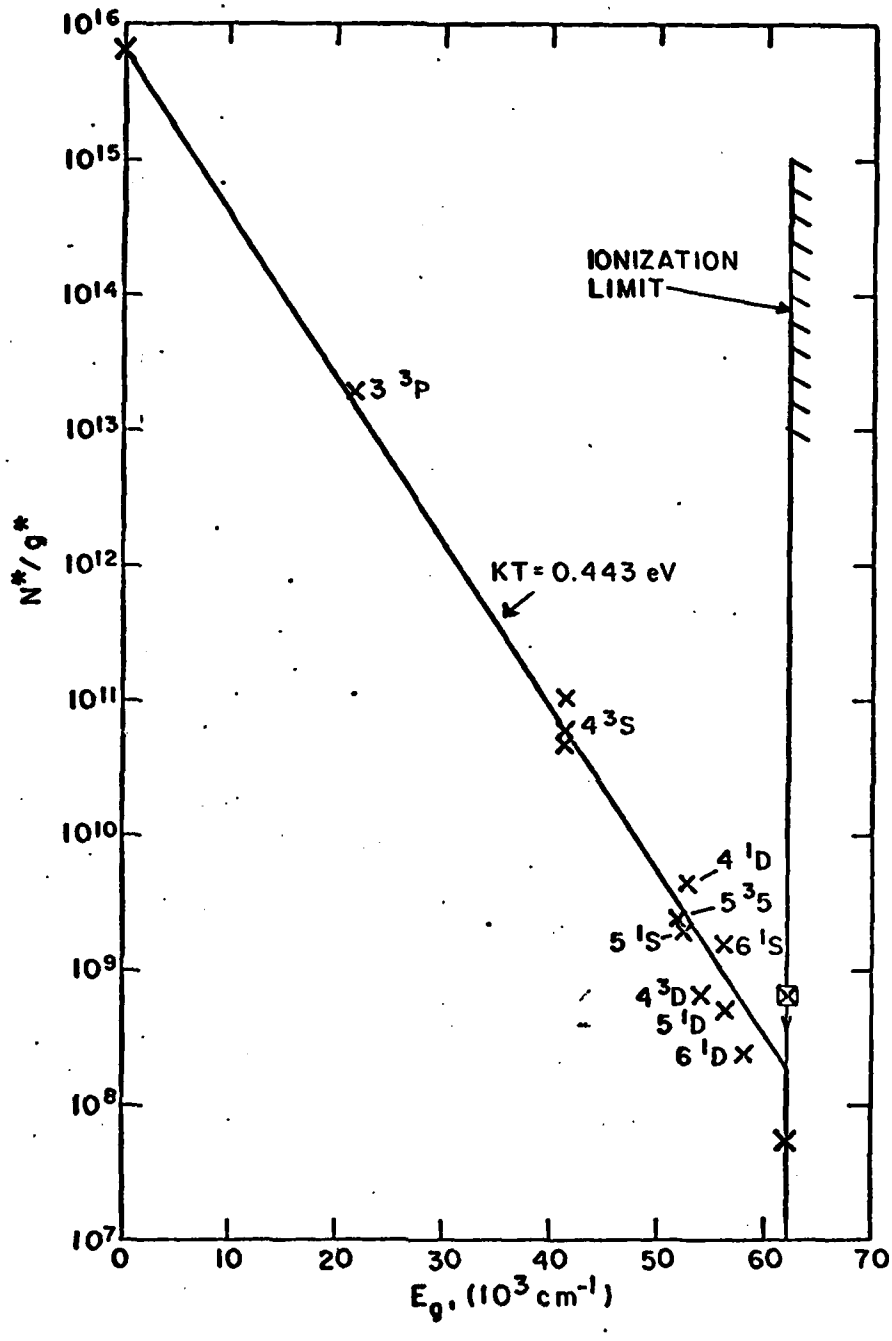

Fig. 2. Typical magnesium-xenon discharge Boltzmann plot. $[Mg]_0 = 7.3 \times 10^{15}/cm^3$, $[Xe] = 2.4 \times 10^{19}/cm^3$, $n_e = 8.3 \times 10^{14}/cm^3$, $E/N = 6.6 \times 10^{-18}$ V-cm^2, $J = 111.6$ A/cm^2.

III. ELECTRON EXCITATION RATES IN METAL VAPORS

Drs. E. R. Mosburg, Jr. and A. V. Phelps

This new experiment is intended to yield effective rate coefficients for electron collisions with highly excited atoms so as to test models of the high power density discharges in metal vapor-rare gas for laser applications. We propose to use a tunable dye laser to modulate the density of selected highly excited levels of mercury atoms in a discharge so as to obtain effective electron collision and ionization rate coefficients for these excited states. The technique of laser perturbation of highly excited state populations has recently been applied by various groups to H, He, Ar^+ and K atoms in low pressure, low electron density discharges.[4] These experiments show that for typical highly excited atomic states and laser pulse lengths (3-10 ns) the maximum electron density for which the excited state transient can be observed is in the 10^{12}-10^{13} cm^{-3} range. Although these electron densities are not much below those used in many current lasers, they are far below the electron densities present in our metal vapor-rare gas discharges. This difference points to the possibility of quasi-steady-state measurements in which the changes in radiation emitted by the perturbed energy level are compared with our theoretical model. A second important difference from many previous experiments is our interest in the change in electron density produced by the irradiation of the excited atoms. Thus, we wish to determine the magnitude of the coupling coefficients between the densities of the various excited states and the density of electrons and to relate this data to the electron loss and production processes, e.g., dissociative recombination and multistep ionization. The importance of dissociative recombination for the high pressure discharges of interest for metal vapor excimer laser

discharges has been discussed in previous Semiannual reports. Thus, instead of the diffusion controlled discharges used in other laser perturbation experiments we will be examining processes relevant to the high power discharges in which electron-ion recombination balances multistep ionization.

IV. ELECTRON EXCITATION OF METASTABLE ATOMS AND MOLECULES

Drs. K. Tachibana and A. V. Phelps

Electron excitation rate coefficients for N_2 are important to the development of models of N_2 lasers,[5] gas discharges[6] and ionospheric phenomena[7] and in determining electron collision cross sections.[8-11] Although cross sections for the excitation of the $N_2(C^3\Pi_u)$ state have been measured by a number of authors,[11] there are only a few measurements of excitation rate coefficients.[12,13] This work presents measurements of this excitation rate coefficient obtained using the drift tube technique used for $O_2(b^1\Sigma_g^+)$ molecules[14] and $N_2(A^3\Sigma_u^+)$ molecules.[15] We also recommend a set of electron-N_2 collision cross sections.

Excitation rate coefficients for the $C^3\Pi_u$ state were obtained from measurements of the absolute intensity of 2nd positive system[16] emitted when electrons drift through N_2 in the presence of an electric field E. The drift tube, detector calibration and data analysis procedures, and corrections for nonuniform detection sensitivity, ionization,[17] etc. are described in Ref. 14. The interference filter was replaced by an f/6.3 monochromator with a resolution (FWHM) of 2.5 nm in order to resolve the 2nd positive and Vegard-Kaplan bands.[16] Most of the measurements were made with 5% H_2 and 95% N_2 in order to maintain electron emission by the gold-paladium photocathode.[15]

The observed band emission is analyzed by first writing a rate equation, e.g., Eq. (3) of Ref. 14, for the density n_i of each vibrational level of the C state. When these equations are added, vibrational relaxation terms cancel.

The radiative transition probabilities are nearly independent of the vibrational level,[16,18] i.e., $A_i = A_{ij} = 2.7 \times 10^7$ sec^{-1}, and diffusion effects[14] are negligible. The assumption that collisional quenching coefficients are independent of the vibrational level leads to a simple relation for the total C-state density, i.e.,

$$n_i = \sum_j \frac{S_{ij}}{g_{ij} A_{ij}} = \frac{(\alpha/N_T)_C \, J_e N_T}{e(A_i + k_q N_T)} . \qquad (1)$$

Here the S_{ij}, g_{ij} and A_{ij} are the signal generated by the detector per unit volume of the source, the detection efficiency, and Einstein coefficient for the transition between the i^{th} vibrational levels of the C state and j^{th} level of the $B^3\Pi_g$ state, $(\alpha/N_T)_C$ is the excitation coefficient per unit distance of electron drift[14]; e and J_e are the electron charge and the average current density; $k_q N_T \equiv k_{qN} N + k_{qH}[H_2]$ is the reciprocal lifetime against quenching by nitrogen k_{qN} and by hydrogen k_{qH}; and N_T, N and $[H_2]$ are the total, nitrogen and hydrogen densities. See Ref. 14 for details of the spatial averaging, etc. The measured S_{ij} were for bands near 337.1(0-0), 315.9(1-0), 297.7(2-0) and 389.5(3-6) nm. Their relative intensities at low N_2 densities agreed well with previous measurements,[13,16] which were used to estimate the small contribution of higher levels. A plot of $J_e N_T (\Sigma n_i)^{-1}$ vs. N_T at $E/N_T = 1 \times 10^{-19}$ V-m^2 yields k_q. Values of $(\alpha/N_T)_C$ were then calculated using Eq. (1). The $(\alpha/N_T)_C$ values were independent of electron current for $1.6 \times 10^{-5} < J_e < 10^{-4}$ A/m^2. The nitrogen density was varied from 3×10^{22} to 6×10^{23} m^{-3}. A measurement in pure N_2 at $E/N = 1 \times 10^{-19}$ V-m^2 yields an $(\alpha/N)_C$ value which is indistinguishable from the corresponding $(\alpha/N_T)_C$ value in the H_2-N_2 mixture. We have therefore dropped the subscript T from the $(\alpha/N_T)_C$ vs. N_T/E data shown in Fig. 3. Empirically, the measured $(\alpha/N)_C$ are fitted by $(6.5 \pm 0.5) \times 10^{-12} \exp[5.5 \times 10^{-19}/(E/N)]$m^2 for $E/N < 2 \times 10^{-19}$ V-m^2.

Our k_q value of 2.1×10^{-17} m^3/sec for 5% H_2 - 95% N_2 is larger than, but consistent with, $k_{qN} = 1.3 \times 10^{-17}$ m^3/sec calculated from pure N_2 data of Legler.[12] It is not consistent with the values for v=0 levels from Urosević et al.[13] or Becker et al.[18] Collision cascading from the E state[19] does not affect our k_q value.

Figure 3 shows that our values of $(\alpha/N)_C$ agree with those measured by Legler.[12] Agreement with the measurements by Urosević et al.[13] is good only at their lowest E/N. The solid curve of Fig. 3 shows our application of the techniques of Ref. 8 to the calculation of $(\alpha/N)_C$ values. The final cross section set included: (a) momentum transfer cross sections from Ref. 8 for electron energies ε below 10 eV and the sum of the elastic and inelastic momentum transfer cross sections of Cartwright[11] for $\varepsilon > 10$ eV; (b) rotational and near-threshold ($\varepsilon < 1.5$ eV) vibrational excitation cross sections from Ref. 8; (c) these vibrational excitation cross sections for $1.7 < \varepsilon < 5$ eV increased by the factor of 1.9 previously found necessary to fit measured excitation rate coefficients for the $A^3\Sigma_u^+$ state[15]; (d) cross sections for the C and E states from Cartwright et al.[11] plus a sharp peak near the E-state threshold[20]; (e) the other electronic excitation cross sections of Cartwright et al. multiplied by 0.8; and (f) ionization cross sections totaling that of Rapp and Englander-Golden.[21] Calculated curves of $(\alpha/N_T)_C$ vs. E/N_T for 5% H_2 - 95% H_2 and of $(\alpha/N)_C$ vs. E/N for pure N_2 differ by less than the experimental scatter. Agreement between calculated and experimental $(\alpha/N)_C$ is good at low E/N. The higher E/N data suggest a need for additional excitation of the C state. Cascading from the E state contributed $\sim$10% to the $(\alpha/N)_C$ values.

The calculated ionization coefficients of Fig. 3 were brought into agreement in magnitude with experiment[17] by applying the 0.8 factor to most

of the excitation cross sections. The more rapid variation of calculated $(\alpha/N)_i$ than experimental $(\alpha/N)_i$ with N/E appears to result from adjusting the vibrational excitation cross section to obtain a fit of calculated and measured A-state excitation coefficients, i.e., earlier calculations[5,8,9] agree with experimental $(\alpha/N)_i$ data but greatly overestimate α/N values for the A state.[15] Efforts to improve the fit between calculation and experiments for N_2 are continuing.[22] In the meantime the present cross section set gives reasonable agreement with five different electron swarm coefficients, i.e., drift velocity,[8] characteristic energy,[8] A-state excitation,[15] C-state excitation and ionization.[17]

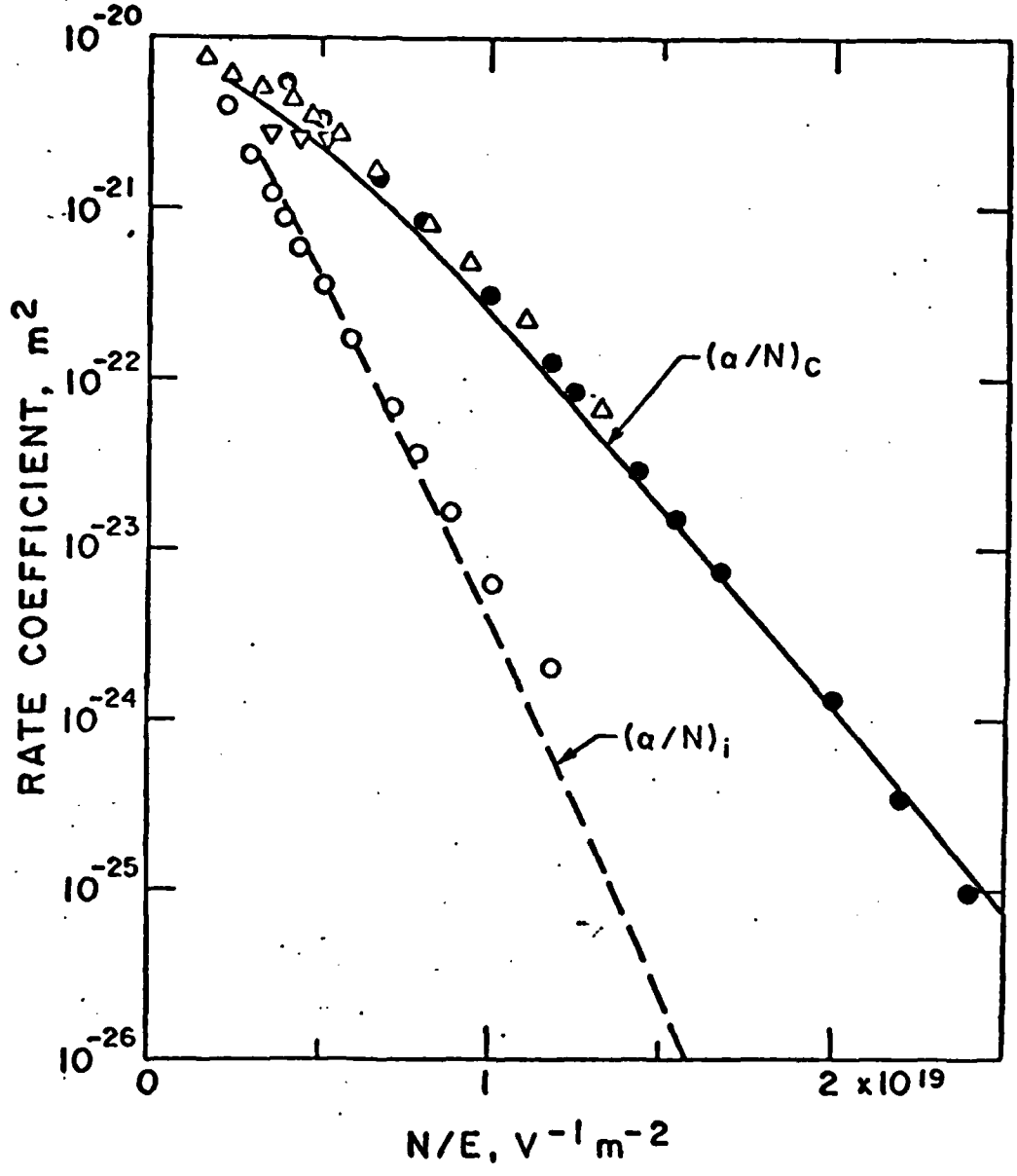

Fig. 3. $C^3\Pi_u$ excitation $(\alpha/N_T)_C$ ionization $(\alpha/N)_i$ coefficients for electrons in N_2. The experimental points and authors are: • this work, Δ -- Legler, ∇ -- Urošević et al., o -- Haydon and Williams. The solid and dashed lines are from calculations using the recommended set of cross sections (see text).

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